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(54) Modified high molecular weight succinimides

(57) Alkenyl or alkyl succinimide additives which are the reaction product of a high molecular weight alkenyl- or alkyl-substituted succinic anhydride and a polyalkylene polyamine having an average of greater than 4 nitrogen atoms per mole, wherein the reaction product is post-treated with a cyclic carbonate, are compatible with fluorocarbon engine seals and, for concentration levels at which fluorocarbon seal compatibility is achieved, possess improved dispersancy and/or detergency properties when employed in lubricating oils and fuels.

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polybutenes, an especially preferred Mn range is 1700-2400. However, the '435 patent also teaches that the succinimides must have a succinic ratio of at least 1.3, that is at least 1.3 succinic groups per equivalent weight of polyalkene-derived substituent group. Most preferred are succinimides having a succinic ratio of 1.5-2.5. The '435 patent teaches that succinimides must have have both a high Mn polyalkylene-derived substituent and a high succinic ratio.

The succinimide additives disclosed in the '435 patent are not only dispersants and/or detergents, but also viscosity index improvers. That is, the '435 additives impart fluidity modifying properties to lubricant compositions containing them. However, viscosity index improving properties are not always desirable for the succinimide, as in the case of single-grade oil formulations, for example.

Polyamino alkenyl or alkyl succinimides and other additives useful as dispersants and/or detergents, such as Mannich bases, contain basic nitrogen. While basicity is an important property to have in the dispersant/detergent additive, it is believed that the initial attack on fluorocarbon elastomer seals used in some engines involves attack by the basic nitrogen. This attack leads to the loss of fluoride ions, and eventually results in cracks in the seals and loss of other desirable physical properties in the elastomer.

One approach towards solving the elastomer problem is described in U.S. Patent No. 4,873,009 to Ronald L. Anderson. This patent is also concerned, in part, with the use of succinimides as lube oil additives. Anderson recognizes in Col. 2, lines 28 et seq. that lube additives prepared from "long chain aliphatic polyamines", i.e., succinimides, "are excellent lube oil additives." Anderson teaches such succinimides are "inferior to additives where the alkylene polyamine is hydroxyalkylated" (Col. 2, lines 31-32). Such hydroxyalkylated polyamine-based succinimides, however, "have the drawback that they tend to attack engine seals particularly those of the fluorocarbon polymer type" (Col. 2, lines 35-37).

Anderson solves his fluorocarbon polymer seal compatibility problem by directly borating his hydroxyalkylated polyamine based succinimides. Furthermore, according to Anderson, it would be desirable for the additive to have a relatively high concentration of N-hydroxyalkyl moieties because the more N-hydroxyalkyl substituents, the cleaner the engine. However, Anderson also teaches that the more amino groups in the polyamine, the greater the degradation of fluorocarbon seal, and that alkylene amines containing more than 2 amino groups cannot be utilized (Col. 2, lines 50-62).

Accordingly, there exists a need in the art for a succinimide lubricating oil additive which is effective in controlling engine sludge and varnish, but which does not require boration to achieve fluorocarbon seal compatibility.

SUMMARY OF THE INVENTION

A unique class of modified polyamino alkenyl or alkyl succinimide compounds has now been found to be simultaneously compatible with fluorocarbon seals and, at concentration levels for which fluorocarbon seal compatibility is achieved, effective in controlling engine sludge and varnish. These modified polyamino alkenyl or alkyl succinimides are prepared from the succinimide reaction product of 1) an alkenyl- or alkyl-substituted succinic anhydride derived from a polyolefin having a Mn of about 2000 to about 2700 and a weight average molecular weight (Mw) to Mn ratio of about 1 to about 5; and 2) a polyalkylene polyamine having greater than 4 nitrogen atoms per mole. The modified succinimides of the present invention are obtained by post-treating the succinimide reaction product with a cyclic carbonate.

Among other factors, the present invention is based on the finding that a unique class of succinimides is effective in controlling engine sludge and varnish at concentration levels for which the succinimides are simultaneously compatible with engine fluorocarbon seals. Generally, known succinimides useful as dispersants and/or detergents are not always compatible with fluorocarbon seals when present in lubricating oil compositions at concentration levels necessary to be effective in controlling engine sludge and varnish. Accordingly, the present invention also relates to a lubricating oil composition containing these modified polyamino alkenyl or alkyl succinimides.

Among other factors, the present invention is also based on the finding that a unique class of modified polyamino alkenyl or alkyl succinimides wherein the alkenyl or alkyl substituent has a Mn in the range of 2000-2700 possess both superior fluorocarbon seal compatibility and superior dispersancy and/or detergency properties compared to those wherein the alkenyl or alkyl substituent has a Mn of less than about 2000.

In addition to lubricating oil compositions, the present invention also relates to fuel compositions comprising a major portion of a hydrocarbon boiling in a gasoline or diesel range and an amount of a modified polyamino alkenyl or alkyl succinimide, compatible with fluorocarbon seals, sufficient to provide dispersancy and/or detergency.

DETAILED DESCRIPTION OF THE INVENTION

The modified polyamino alkenyl or alkyl succinimides of this invention are prepared by post-treating a polyamino alkenyl or alkyl succinimide with a cyclic carbonate. The polyamino alkenyl or alkyl succinimides are typically prepared by reaction of an alkenyl or alkyl succinic anhydride with a polyamine.

Alkenyl or alkyl succinimides are disclosed in numerous references and are well known in the art. Certain funda-

M_{po} = number average molecular weight of the starting polyolefin

M_{ma} = 98 (molecular weight of maleic anhydride)

C = conversion factor = 112220 (for conversion of gram-moles of alkenyl or alkyl succinic anhydride per gram of sample to milligrams of KOH per gram of sample)

The saponification number, P, can be measured using known procedures, such as the procedure described in ASTM D94.

The actives fraction of the alkenyl or alkyl succinic anhydride can be determined from the percent of unreacted polyolefin according to the following procedure. A 5.0 gram sample of the reaction product of maleic anhydride and polyolefin is dissolved in hexane, placed in a column of 80.0 grams of silica gel (Davisil 62, a 140 angstrom pore size silica gel), and eluted with 1 liter of hexane. The percent unreacted polyolefin is determined by removing the hexane solvent under vacuum from the eluent and weighing the residue. Percent unreacted polyolefin is calculated according to the following formula:

$$\text{Percent Unreacted Polyolefin} = \frac{\text{Net Weight of Residue}}{\text{Sample Weight}} \times 100$$

The weight percent actives for the alkenyl or alkyl succinic anhydride product is calculated from the percent unreacted polyolefin using the formula:

$$\text{Weight Percent Actives} = 100 - \text{Percent Unreacted Polyolefin}$$

The actives fraction of the alkenyl or alkyl succinic anhydride is then calculated as follows:

$$\text{Actives Fraction} = \frac{\text{Weight Percent Actives}}{100}$$

The percent conversion of polyolefin is calculated from the weight percent actives as follows:

$$\text{Percent Conversion} = \frac{\text{wt. \% actives} \times \left[\frac{M_{po}}{M_{po} + [M_{ma} \times SR]}\right]}{\left[\text{wt. \% actives} \times \left[\frac{M_{po}}{M_{po} + [M_{ma} \times SR]}\right]\right] + [100 - \text{wt. \% actives}]}$$

wherein

M_{po} = number average molecular weight of the starting polyolefin

M_{ma} = 98 (molecular weight of maleic anhydride)

SR = succinic ratio of alkenyl or alkyl succinic anhydride product

It is, of course, understood that alkenyl or alkyl succinic anhydride products having high succinic ratios can be blended with other alkenyl succinic anhydrides having lower succinic ratios, for example, ratios of around 1.0, to provide an alkenyl succinic anhydride product having an intermediate succinic ratio.

In general, suitable succinic ratios for the alkenyl or alkyl succinic anhydride reactants employed in preparing the additives of this invention are greater than about 1 but less than about 2. Succinic anhydrides with succinic ratios of about 2, when reacted with amines having greater than 4 nitrogen atoms per mole and post-treated with a cyclic carbonate, form gels. Accordingly, succinic ratios of about 1.7 or less are preferred.

The Polyamino Reactant

The polyamine to be reacted with the alkenyl or alkyl succinic anhydride in order to produce the polyamino alkenyl

the number of moles of polyamine to the number of moles of succinic groups in the succinic anhydrid reactant. The number of moles of succinic groups in the succinic anhydride reactant is determined as follows:

$$\text{number of moles of succinic groups} = \frac{P}{C} \times \text{weight of alkenyl or alkyl succinic anhydride sample (g)}$$

wherein P and C are as defined above.

POST-TREATMENT OF THE POLYAMINO ALKENYL OR ALKYL SUCCINIMIDE WITH A CYCLIC CARBONATE

The polyamino alkenyl or alkyl succinimides formed as described above are then reacted with a cyclic carbonate. The resulting modified polyamino alkenyl succinimide has one or more nitrogens of the polyamino moiety substituted with a hydroxy hydrocarbyl oxycarbonyl, a hydroxy poly(oxyalkylene) oxycarbonyl, a hydroxyalkylene, hydroxyalkylenepoly(oxyalkylene), or mixture thereof. The products so produced are compatible with fluorocarbon seals and are effective dispersant and detergent additives for lubricating oils and for fuels.

The reaction of a polyamino alkenyl or alkyl succinimide with a cyclic carbonate is conducted at a temperature sufficient to cause reaction of the cyclic carbonate with the polyamino alkenyl or alkyl succinimide. In particular, reaction temperatures of from about 0°C to about 250°C are preferred with temperatures of from about 100°C to 200°C being more preferred and temperatures of from 150°C to 180°C are most preferred.

The reaction may be conducted neat, wherein both the alkenyl or alkyl succinimide and the cyclic carbonate are combined in the proper ratio, either alone or in the presence of a catalyst (such as an acidic, basic or Lewis acid catalyst), and then stirred at the reaction temperature. Examples of suitable catalysts include, for instance, phosphoric acid, boron trifluoride, alkyl or aryl sulfonic acid, alkali or alkaline carbonate.

Alternatively, the reaction may be conducted in a diluent. For example, the reactants may be combined in a solvent such as toluene, xylene, oil or the like, and then stirred at the reaction temperature. After reaction completion, volatile components may be stripped off. When a diluent is employed, it is preferably inert to the reactants and products formed and is generally used in an amount sufficient to insure efficient stirring.

Water, which can be present in the polyamino alkenyl or alkyl succinimide, may be removed from the reaction system either before or during the course of the reaction via azeotroping or distillation. After reaction completion, the system can be stripped at elevated temperatures (100°C to 250°C) and reduced pressures to remove any volatile components which may be present in the product.

Alternatively, a continuous system may be employed in which the alkenyl or alkyl succinic anhydride and polyamine are added at the front end of the system while the organic carbonate is added further downstream in the system. In such a continuous system, the organic carbonate may be added at any time after mixing of the alkenyl or alkyl succinic anhydride with the polyamine has occurred. Preferably, the organic carbonate is added within two hours after mixing of the alkenyl or alkyl succinic anhydride with the polyamine, preferably after the major portion of the amine has reacted with the anhydride.

In a continuous system, the reaction temperature may be adjusted to maximize reaction efficiency. Accordingly, the temperature employed in the reaction of the alkenyl or alkyl succinic anhydride with a polyamine may be the same as or different from that which is maintained for the reaction of this resulting product with the cyclic carbonate. In such a continuous system, the reaction temperature is generally between 0°C to 250°C; preferably between 125°C to 200°C; and most preferably between 150°C to 180°C.

The reaction of polyamino alkenyl or alkyl succinimides with cyclic carbonates is known in the art and is described in U.S. Patent 4,612,132, which is totally incorporated herein by reference.

A particularly preferred cyclic carbonate is 1,3-dioxolan-2-one (ethylene carbonate). Ethylene carbonate is commercially available or may be prepared by methods well-known in the art.

The molar charge of cyclic carbonate employed in the post-treatment reaction is based upon the theoretical number of basic nitrogens contained in the polyamino substituent of the succinimide. Thug, when 1 equivalent of tetraethylene pentamine ("TEPA") is reacted with two equivalents of succinic anhydride, the resulting bis succinimide will theoretically contain 3 basic nitrogens. Accordingly, a molar charge of 2 would require that two moles of cyclic carbonate be added for each basic nitrogen or in this case 6 moles of cyclic carbonate for each mole of bis succinimide prepared from TEPA. Mole ratios of the cyclic carbonate to the basic amine nitrogen of the polyamino alkenyl succinimide employed in the process of this invention are generally in the range of from about 1.5:1 to about 4:1; although preferably from about 2:1 to about 3:1.

As described in U.S. Patent No. 4,612,132, cyclic carbonates may react with the primary and secondary amines of a polyamino alkenyl or alkyl succinimide to form two types of compounds. In the first instance, strong bases, including unhindered amines such as primary amines and some secondary amines, react with an equivalent of cyclic carbonate to produce a carbamic ester. In the second instance, hindered bases, such as hindered secondary amines, may react with an equivalent of the same cyclic carbonate to form a hydroxyalkyleneamine linkage. Unlike the carbamate prod-

in the base fuel is 10 to 10,000 weight parts per million, preferably from 30 to 2,000 weight parts per million, of the modified succinimide per part of base fuel. If other detergents are present, a lesser amount of the modified succinimide may be used.

The modified succinimide additives of this invention may also be formulated as a fuel concentrate, using an inert stable oleophilic organic solvent boiling in the range of about 150°F to 400°F. Preferably, an aliphatic or an aromatic hydrocarbon solvent is used, such as benzene, toluene, xylene or higher-boiling aromatics or aromatic thinners. Aliphatic alcohols of about 3 to 8 carbon atoms, such as isopropanol, isobutylcarbinol, n-butanol and the like, in combination with hydrocarbon solvents are also suitable for use with the fuel additive. In the fuel concentrate, the amount of the additive will be ordinarily at least 10 percent by weight and generally not exceed 70 percent by weight and preferably from 10 to 25 weight percent (all on a dry polymer basis).

The following examples are offered to specifically illustrate this invention. These examples and illustrations are not to be construed in any way as limiting the scope of this invention.

EXAMPLES

Example 1. Preparation of PIBSA 2200 (succinic ratio = 1.1)

A 35.186 Kg, 16 mol., sample of Parapol 2200 (a 2200 Mn polybutene available from Exxon Chemical Company) was charged to a reactor and heated to 232°C. During this time, the reactor was pressurized to 40 psig with nitrogen and then vented three times to remove oxygen. The reactor was pressurized to 24.7 psia. Then 1500 g maleic anhydride was added over a thirty-minute period. Then 4581 g maleic anhydride was added over a 4-hour period. The total charge mole ratio (CMR) of maleic anhydride to polybutene was 3.88. After the maleic anhydride addition was completed, the reaction was held at 232°C for 1.5 hour. Then the reaction was cooled and the pressure reduced to 0.4 psia to remove any unreacted maleic anhydride. To this was then added a light neutral diluent oil. This was heated to 160°C for 24 hours and was then filtered. This product was found to contain 37.68 wt. % actives and had a saponification number of 19.7 mg KOH/g sample. The succinic ratio was 1.1 based on a polybutene molecular weight of 2246 determined by GPC.

Example 2. Preparation of PIBSA 1300 (succinic ratio = 1.1)

The procedure of Example 1 was repeated except that Parapol 1300 (a 1300 Mn polybutene available from Exxon Chemical Company) was used instead of Parapol 2200. After dilution with diluent oil and filtration, this product was found to contain 49.6 wt. % actives and a saponification number of 42.2 mg KOH/g sample. The succinic ratio was 1.1 based on a polybutene molecular weight of 1300.

Example 3. Preparation of PIBSA 2200 (succinic ratio = 1.5)

Parapol 2200, 42.8 Kg, 19.45 mol, was charged to a reactor and the temperature was increased to 150°C. During this time, the reactor was pressurized to 40 psig with nitrogen and then vented three times to remove oxygen. Then at 150°C, maleic anhydride, 4294 g, 43.82 mol, and di-t-butylperoxide, 523 g, 3.58 mol, was added. The first 25% was added over 30 minutes. The remainder was then added over 11.5 hours. The CMR of maleic anhydride to polybutene was 2.25. The reaction was held at 150°C for one hour. Then the reactor was heated to 190°C for 1 hour to destroy any remaining di-t-butylperoxide. Then vacuum was applied to the reactor and the unreacted maleic anhydride was removed. This material was then diluted with a light neutral oil and filtered. The product after filtration had a saponification number of 31.6 mg KOH/g sample and contained 45.62 wt. % actives. The succinic ratio was 1.5 for this material based on a polybutene molecular weight of 2200.

Example 4A. Preparation of PIBSA 1300 (succinic ratio = 1.9)

Parapol 1300, 6.9 Kg, 47.6 mol, was charged to a reactor and the temperature was increased to 150°C. During this time, the reactor was pressurized to 40 psig with nitrogen and then vented three times to remove oxygen. Then at 150°C, maleic anhydride, 9332.66 g (95.23 mol), and di-t-butylperoxide, 1280 g (8.77 mol) was added over 5 hours. Then the reaction was maintained at 150°C for an additional 2 hours. The reaction was then heated to 190°C for 1 hour to destroy any residual peroxide. The pressure was then reduced to 0.4 psia and the excess maleic anhydride was removed. The product was found to contain 65.4 wt. % actives and had a saponification number of 94.5 mg KOH/g sample. The succinic ratio was 1.9 for this material based on a polybutene molecular weight of 1300.

TABLE I

(Analytical Data For Examples 5-19)

		MEASURED			
Compound of Example No.:	**DESCRIPTION**	**%N**	**TBN**	**VIS 100 *(cSt)***	**SpGr *(15°C)***
5	bis HPA-X PIBSA 2200 (SR=1.1; A/P=0.44)	0.74	17	428	0.9106
6	bis TETA PIBSA 1300 (SR=1.1; A/P=0.5)	0.99	15	278	0.9300
7	bis HPA-X PIBSA 2200 (SR=1.5; A/P=0.5)	1.05	25	1688	0.9219
8	bis HPA-X PIBSA 1300 (SR=1.1; A/P=0.5)	1.55	36	272	0.9214
9	bis TETA PIBSA 2200 (SR=1.5; A/P=0.5)	0.64	10	1554	0.9339
10	bis TETA PIBSA 2200 (SR=1.1; A/P=0.44)	0.41	5	491	0.9093
11	EC bis HPA-X PIBSA 1300 (SR=1.1; A/P=0.5; EC/BN=2.0)	1.51	20	447	0.9393
12	EC bis TETA PIBSA 1300 (SR=1.5; A/P=0.5; EC/BN=2.0)	0.96	8	305	0.9282
13	bis TETA PIBSA 1300 (SR-1.5; A/P=0.5)	0.87	15	145	0.9120
14	bis HPA-X PIBSA 1300 (SR=1.5; A/P=0.5)	1.52	37	165	0.9142
15	EC bis TETA PIBSA 1300 (SR-1.5; A/P=0.5; EC/BN=2.0)	0.99	11	136	0.9156
16	EC bis HPA-X PIBSA 1300 (SR=1.5; A/P=0.5; EC/BN=2.0)	1.46	19	402	0.9330
17	EC bis HPA-X PIBSA 2200 (SR-1.1; A/P=0.44; EC/BN=2.0)	0.63	9	660	0.9188
18	EC bis HPA-X/DETA PIBSA 2200 (SR=1.1; A/P=0.40; EC/BN=2.4)	0.44	6	485	0.9132
19	EC bis HPA-X/DETA PIBSA 1300 (SR-1.1; A/P=0.5; EC/BN=2.0)	1.18	9.7	287	

Note:
SR = succinic ratio
A/P = amine/PIBSA CMR
EC/BN = ethylene carbonate/basic nitrogen CMR

Blending of Samples on an Equal Basis

We chose to blend and test the additives in Examples 5-19 on an equal wt. % actives basis. This was because we were trying to compare products from four different PIBSA's with different molecular weights and different succinic ratios, and two different amines with and without ethylene carbonate treatment. In order to do this, we calculated the %N and TBN that was expected for these compounds from the molecular formulas for a product that contained 40 wt. % actives. These data are reported in Table II. The succinimides from Examples 5-18 were then blended into the finished oil for testing at a concentration of 7.5% of the 40 wt. % actives material or at 3% on a dry polymer basis. The amounts of succinimides were adjusted to take into account the differences between the %N of the particular batch and the %N expected for the example. For Example 19, a 5% blend of 50 wt. % actives material or 3% on a dry polymer basis was made.

TABLE III - (PV-3344 TEST RESULTS)

Additive Compound of Example No.	Concentration of Additive (Wt. %)	TS (Pass ≥ 8.0)	EL (Pass ≥ 160)	CR (Pass = N)
5	1.6	10.0	203	N
	2.0	9.4	189	N
	2.4	8.8	196	N
	2.4	8.0	175	Y
	2.8	7.8	176	Y
	3.2	7.2	167	Y
6	1.6	10.8	218	N
	2.4	9.6	197	N
7	1.6	10.9	220	N
8	1.6	6.5	155	Y
	2.4	6.0	146	Y
9	1.6	11.7	232	N
10	1.6	12.5	244	N
	3.2	11.7	240	N
11	1.6	6.0	139	Y
	2.8	5.8	141	Y
12	1.6	10.9	216	N
13	1.6	11.2	224	N
	2.4	9.4	196	N
14	1.6	6.9	160	Y
	2.4	5.6	137	Y
15	1.6	11.7	233	N
	2.4	10.7	207	N
16	1.6	6.8	153	Y
	2.4	6.4	148	Y
17	1.6	9.0	188	N
	2.0	8.8	180	N
	2.4	8.8	196	N
	2.8	7.5	172	Y
	3.2	7.9	169	Y
18	1.6	12.1	238	N
	2.0	11.6	233	N
	2.4	11.1	220	N
	2.8	10.7	220	N
	3.2	10.0	206	N
19	1.6	10.1	186	N
	2.8	8.3	150	Y

The detergency properties of the additive compounds were then tested using the Sequence VE engine test procedure, as defined in ASTM Proposed Method:212. This test measures, among other things, average engine sludge (AES) and average engine varnish (AEV). The AES and AEV results for the compounds of Examples 5-19 are shown in Table IV. A dosage or treat rate level of 3.0% (on a dry polymer basis) was chosen as an appropriate concentration level for the Seq. VE test since treat rate levels exceeding 3% are generally too high for the resulting additive package to be priced competitively in the marketplace. Examples 17 and 18 were each run at concentration levels of 2.0 and 1.5% (on a dry polymer basis).

TABLE V (continued)

(EFFECT OF POLYBUTENE Mn)

Compound of Example No.:	Succinic Ratio	Amine Type	Ethylene Carbonate Post-Treatment	Polybutene Mn	PV-3344 TS	Seq. VE AES	Seq. VE AEV
8	1.1	HPA-X	No	1300	6.5	7.7	4.6
5	1.1	HPA-X	No	2200	10.0	9.4	5.6
11	1.1	HPA-X	Yes	1300	6.0	9.1	5.9
17	1.1	HPA-X	Yes	2200	9.0	9.4	5.9
14	1.5	HPA-X	No	1300	6.9	9.3	5.4
7	1.5	HPA-X	No	2200	10.9	9.5	6.0
13	1.5	TETA	No	1300	11.2	9.1	5.1
9	1.5	TETA	No	2200	11.7	9.3	5.6
Average	-	-	-	1300	8.3	8.6	4.9
Average	-	-	-	2200	10.8	9.3	5.4

Table V demonstrates that a polyisobutene Mn of 2200 gives better PV-3344 and better Seq. VE results than a polyisobutene Mn of 1300.

TABLE VI

(EFFECT OF AMINE TYPE)

Compound of Example No.:	Polybutene Mn	Succinic Ratio	Ethylene Carbonate Post-Treatment	Amine Type	PV-3344 TS	Seq. VE AES	Seq. VE AEV
6	1300	1.1	No	TETA	10.8	8.0	3.4
8	1300	1.1	No	HPA-X	6.5	7.7	4.6
10	2200	1.1	No	TETA	12.5	8.9	4.0
5	2200	1.1	No	HPA-X	10.0	9.4	5.6
9	2200	1.5	No	TETA	11.7	9.3	5.6
7	2200	1.5	No	HPA-X	10.9	9.5	6.0
12	1300	1.1	Yes	TETA	10.9	8.7	4.1
11	1300	1.1	Yes	HPA-X	6.0	9.1	5.9
13	1300	1.5	No	TETA	11.2	9.1	5.1
14	1300	1.5	No	HPA-X	6.9	9.3	5.4
15	1300	1.5	Yes	TETA	11.7	9.4	5.3
16	1300	1.5	Yes	HPA-X	6.8	9.4	6.4
Average	-	-	-	TETA	11.5	8.9	4.6
Average	-	-	-	HPA-X	7.9	9.1	5.6
17	2200	1.1	Yes	HPA-X	9.0	9.4	5.9

TABLE VIII

(CONCLUSIONS)			
	Better PV-3344 Performance	**Better Seq. VE (AES) Performance**	**Better Seq. VE (AEV) Performance**
A. Polyisobutene Mn (1300 or (2200)	2200	2200	2200
B. Post-Treatment (Yes or No) with ethylene carbonate	No (slightly)	Yes	Yes
C. Amine type 1. TETA or HPA-X	TETA	HPA-X (slightly)	HPA-X
2. HPA or DETA/HPA-X	DETA/HPA-X	HPA-X (slightly)	HPA-X

Table VIII shows that the most desirable additives contain a 2200 Mn substituent, are derived from a polyamine having greater than 4 nitrogen atoms per mole, and are post-treated with ethylene carbonate.

While TETA appears to be the best amine type for PV-3344 performance, the concentration levels required for this amine type to achieve suitable Seq. VE performance (AEV results in particular) are unacceptable because they are too high to allow for a competitive treat rate. Accordingly, the amine should have greater than 4 nitrogen atoms per mole.

For multi-grade oil applications, the succinimide additive may be derived from a succinic anhydride having a succinic ratio of approximately 1.5. However, the viscosity index improvement which accompanies succinimides having succinic ratios of about 1.3 or greater is not always desirable. Instead, for some applications, such as single-grade oil formulation, a succinic ratio less than about 1.3, preferably closer to 1, is more desirable. Furthermore, Example 20 (made from the PIBSA of Example 4A) shows that succinic ratios of about 1.9 are unacceptable because gels are formed. Accordingly, succinic ratios greater than 1 but less than about 2 are acceptable, with succinic ratios less than about 1.7 preferred.

Succinimide additives having a 2200 Mn alkenyl or alkyl group which are derived from an amine having greater than 4 nitrogen atoms per mole, and which are post-treated with ethylene carbonate, are compatible with fluorocarbon seals at concentration levels for which they are excellent detergent additives. Such additive compounds (Examples 17 and 18) pass the Seq. VE test at low concentration levels and are desirable because less of the additive is needed in additive packages, thereby resulting in lower-cost oil formulations.

Claims

1. A lubricating oil composition comprising a major amount of an oil of lubricating viscosity and an effective amount of a modified polyamino alkenyl or alkyl succinimide sufficient to be compatible with fluorocarbon seals and simultaneously control engine sludge and varnish, wherein the modified succinimide comprises the succinimide reaction product of:

   (i) an alkenyl- or alkyl-substituted succinic anhydride derived from a polyolefin having a Mn of about 2000 to about 2700 and a Mw/Mn ratio of about 1 to about 5; and
   (ii) a polyalkylene polyamine having greater than 4 nitrogen atoms per mole;
   wherein the succinimide reaction product is post-treated with a cyclic carbonate.

2. A lubricating oil composition according to Claim 1 wherein the charge mole ratio of (ii) to (i) is from about 0.35:1 to about 0.6:1; and the charge mole ratio of cyclic carbonate to basic amine nitrogen in the succinimide reaction product is from about 1.5:1 to about 4:1.

3. A lubricating oil composition according to Claim 1 wherein the polyolefin has a Mn of about 2100 to about 2400.

4. A lubricating oil composition according to Claim 3 wherein the polyolefin has a Mn of about 2200.

5. A lubricating oil composition according to Claim 1 wherein the polyolefin is polybutene.

cyclic carbonate is ethylene carbonate, and the charge mole ratio of cyclic carbonate to basic amine nitrogen in the succinimide reaction product is from about 2:1 to about 3:1.

21. A lubricating oil concentrate according to Claim 19 wherein the polyolefin is polyisobutene having a Mn of about 2200, the succinic anhydride has a succinic ratio from about 1 to about 1.7, the polyalkylene polyamine comprises 20% by weight diethylene triamine and 80% by weight Union Carbide HPA-X heavy polyamine, the charge mole ratio of (ii) to (i) is from about 0.4:1 to about 0.5:1, the cyclic carbonate is ethylene carbonate, and the charge mole ratio of cyclic carbonate to basic amine nitrogen in the succinimide reaction product is from about 2:1 to about 3:1.

22. A fuel composition comprising a hydrocarbon boiling in the gasoline or diesel range and from about 10 to about 10,000 weight parts per million on a dry polymer basis of a modified polyamino alkenyl or alkyl succinimide comprising the succinimide reaction product of:

(i) an alkenyl- or alkyl-substituted succinic anhydride derived from a polyolefin having a Mn of about 2000 to about 2700 and a Mw/Mn ratio of about 1 to about 5; and
(ii) a polyalkylene polyamine having greater than 4 nitrogen atoms per mole;
wherein the succinimide reaction product is post-treated with a cyclic carbonate.

23. A fuel concentrate comprising an inert stable oleophilic organic solvent boiling in the range of 150°F to 400°F and from about 10 to about 70 weight percent on a dry polymer basis of a modified polyamino alkenyl or alkyl succinimide comprising the succinimide reaction product of:

(i) an alkenyl- or alkyl-substituted succinic anhydride derived from a polyolefin having a Mn of about 2000 to about 2700 and a Mw/Mn ratio of about 1 to about 5; and
(ii) a polyalkylene polyamine having greater than 4 nitrogen atoms per mole;
wherein the succinimide reaction product is post-treated with a cyclic carbonate.